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<u>L4</u>	(releas\$3 or permeab\$9 near5 electro\$8) near5 cell\$1	25480	<u>L4</u>
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File: USPT

Nov 9, 1999

DOCUMENT-IDENTIFIER: US 5983131 A

TITLE: Apparatus and method for electroporation of tissue

Abstract Text (1):

The present invention relates to an apparatus and method for localized electroporation of tissue. An apparatus includes a perforate electrically insulating layer, a first electrode at a first side of the perforate electrically insulating layer and a second electrode at a second side of the perforate electrically insulating layer. An electric field extending between the first and second electrodes will preferentially extend through perforations of the electrically insulating layer. The electric field thereby causes electroporation of tissue that is proximate to the first electrode and is partitioned from the electrically insulating layer and the second electrode by the first electrode. The apparatus controllably limits the depth of the electric field within a tissue, such as skin, thereby electroporating a surface layer, such as a stratum corneum layer of the skin, without stimulating submerged nerve endings within the skin. Sensation of the applied voltage and especially pain sensation can thereby be controlled by the invention.

Brief Summary Text (3):

One attempt to solve problems presented by transfer of molecules and ions across tissues, in particular skin tissue, is employment of a phenomenon called electroporation. Generally, electroporation is a method of temporarily or permanently increasing the permeability of tissue and cell membranes, and simultaneously providing an electrical driving force. The increased permeability allows transport or migration, of chemical agents through the tissue or across cell membranes into cells. Electroporation has been used to deliver drugs to tissue, in vivo, by applying electrodes to the surface of an organism and applying a voltage between the electrodes which exposes the tissue to an electric field. The tissue becomes electroporated and allows delivery of a chemical agent, such as a drug, which has been applied either topically to the organism or injected into the bloodstream of the organism, across the electroporated tissue and into cells of the electroporated tissue.

Brief Summary Text (4):

The effect of electroporation on tissue can be temporary or long-lasting. Without continued application of an electric field, electroporated tissue often reverts back to its original condition. However, the duration of electroporation is dependent upon the degree of electroporation of the tissue. In other words, to obtain a longer duration of electroporation, the period of time of applied voltage, or the amount of voltage applied, must be increased. However, discharge of electrical pulses to skin usually causes sensation by the individual receiving treatment. That sensation is caused by applying an electrical voltage to skin is well known. Often, to create a period of electroporation that is sufficient to deliver a therapeutic amount of a chemical agent, the period of pulsation and/or the required voltage can be uncomfortable or even painful to a patient.

Brief Summary Text (8):

The apparatus includes a perforate electrically insulating layer, a first electrode at a first side of the perforate electrically insulating layer and a second electrode at a second side of the perforate electrically insulating layer whereby an electric field extending between the first and second electrodes will preferentially extend through perforations of the electrically insulating layer. The electric field thereby causes electroporation of a tissue that is proximate to the first electrode and is partitioned from the electrically insulating layer and the second electrode by the first electrode.

Brief Summary Text (9):

In one embodiment, the apparatus includes an outer wall and an inner electrically-insulating wall. The inner electrically-insulating wall is recessed within the outer wall. The inner and outer walls define first and second reservoirs. A first electrode is at said first reservoir and a second electrode is at said second reservoir, whereby an electric field extending between electrically-conductive compositions within said first and second reservoirs will extend around the inner electrically-insulating wall and through tissue, at said outer and inner walls, that is in contact with the electrically conductive compositions, the electric field thereby causing electroporation of tissue that is proximate to the electrically conductive compositions.

Brief Summary Text (10):

The method includes applying at least one first electrode to the tissue and forming an electric field between the first electrode and a second electrode that is partitioned from the first electrode by a nonconductive layer. The electric field extends around at least a portion of the insulating layer, whereby the electric field causes electroporation of tissue that is proximate to the first electrode.

Brief Summary Text (11):

The apparatus of the invention has several advantages. For example, an electric field generated between the first and second electrodes will preferentially be limited to perforations of the electrically insulating layer and portions of tissue, such as skin, most proximate to those perforations. Further, electric field lines that extend from one electrode at the tissue to another electrode located away from the tissue will generally limit the depth of the electric field lines within the tissue. This is particularly advantageous for skin tissue, wherein nerve endings lie between about 75 and about 100 micrometers below the surface layer, or stratum corneum, of the skin. Limiting the depth of electric field lines by use of the apparatus of the invention enables electroporation of the stratum corneum without stimulating nerve endings, thereby substantially preventing sensation, including pain, and also preventing stimulation of muscle and associated tissue movement. In other embodiments, the electric field lines can be controlled by, for example, adjusting the amount of applied voltage to the electrodes, to thereby control the amount of sensation during electroporation.

Drawing Description Text (6):

FIG. 5 is a side view, shown in cross-section, of an embodiment of the invention having a plurality of first electrodes separated by an electrically insulating layer.

Drawing Description Text (8):

FIG. 7 is a plot of electrical resistance of epidermis of one subject versus pulse voltage applied to the epidermis by the method of the invention.

Drawing Description Text (9):

FIG. 8 is a plot of electrical resistance of epidermis of another subject versus pulse voltage applied to the epidermis by the method of the invention.

Drawing Description Text (10):

FIG. 9 is a plot of electrical resistance versus time following application of an electrical pulse to epidermis by the method of the invention.

Detailed Description Text (3):

Generally, electroporation is a method of increasing the permeability of tissue and cell membranes and simultaneously providing a driving force for molecular transport. The increased permeability allows transport, or migration, of chemical agents through the tissue or across cell membranes into cells. For example, electroporation can include applying a voltage across tissue in vitro to cause the permeability of the tissue and cell membranes of cells in the tissue to increase. If the tissue is in the presence of a suitable chemical agent, the chemical agent can then migrate across the tissue or into cells of the tissue. Electroporation has also been used to deliver drugs to tissue, in vivo, by applying electrodes to the surface of an organism and applying a voltage between the electrodes which exposes the tissue to an electric field. The tissue thereby becomes electroporated and allows delivery of a chemical agent, such as a drug, which has been applied either topically to the organism or injected into the blood stream of the organism, across the electroporated tissue and

into cells of the electroporated tissue.

Detailed Description Text (4):

"Electroporation," as that term is used herein, means increased permeability of a cell membrane and/or at least a portion of cells of a targeted tissue to a chemical agent or other suitable material, wherein the increased permeability is caused by application of voltage across the cell or at least a portion of the tissue. An example of a suitable target tissue would be skin tissue, or epidermis, including the multilamellar bilayer membrane within the stratum corneum and also envelopes of keratinocytes. The chemical agent can thereby migrate into or across the electroporated tissue and/or across the cell membrane and into the cell.

Detailed Description Text (5):

Examples of apparatus that have been used for electroporation are disclosed in U.S. Ser. No. 07/705,778, filed May 28, 1991 and in U.S. Pat. No. 5,091,034, issued May 28, 1991, the teachings of which are incorporated by reference in their entirety. Other methods can also be employed, such as those described in U.S. Patent Application entitled, "Introduction of Modifying Agents into Skin by Electroporation," by James C. Weaver, Thomas E. Zewert, Uwe Pliquett, Rita Vanbever, Mark R. Prausnitz, Tani Chen, Christopher Cullander, Richard Guy and Robert S. Langer, Ser. No. 08/695,367, pending, the teachings of which are incorporated herein in their entirety. For example, electric pulses generated by suitable apparatus to cause electroporation by the method of the invention typically are exponential pulses having a time constant in the range of between about 0.1 and about 3 milliseconds, but in some cases as long as about 300 milliseconds, and an amplitude in the range of between about 30 and 1000 volts across the electrodes, which results in smaller voltages across the skin. However, the pulse configuration can also be square, bipolar, etc. Generally, the number of pulses sufficient to cause electroporation is in the range of between about 1 and about 10, wherein the interval between pulses is in the range of between about 0.01 second and 1 minute. Usually the largest increase in permeability caused by electroporation occurs as a consequence of applying the first pulse.

Detailed Description Text (6):

In one embodiment, the apparatus represented in FIG. 1 can be employed. As shown in FIG. 1, electrode/reservoir device 10 is located at epidermis 12 of a human patient. Electrode/reservoir device 10 includes housing 14 that defines application opening 16 and inlet 18. Housing 14 is suitable for containing composition 20, at least one component of which is to be administered to the patient. First electrode 22 is located at application opening 16 within housing 14 and contacts epidermis 12. As shown in FIG. 1, first electrode 22 is a perforate plate. Perforations 24 in first electrode 22 typically have a diameter in a range of between about 5 .mu.m and 500 .mu.m, and, preferably, in a range of between about 10.mu. and 50 .mu.m. However, first electrode 22 can be alternatively, a bar, a wire mesh, a plate with slit-shaped holes, or have some other configuration. Perforate electrically-insulating layer 26 is located above first electrode 22.

Detailed Description Text (7):

In the case of skin tissue, electrode/reservoir device 10 causes the electric field to be mostly confined to the stratum corneum, i.e., minimal penetration of the electric field into the viable epidermal tissue adjacent the stratum corneum. Accordingly, the diameter of the hole(s) in first electrode 22 is generally chosen to be larger than the diameter of the hole(s) in electrically insulating layer 26. For example, if a 50 .mu.m diameter hole is used in electrically insulating layer 26, the hole diameter is first electrode 22 can be about 80 .mu.m, which provides an annular gap between the two holes of about 15 .mu.m. Generally the width of the annular gap should be approximately equal to the thickness of the stratum corneum. Thus, for human skin with a stratum corneum thickness of about 20 .mu.m, the width of the annular gap should be in a range of between about 1 .mu.m and about 50 .mu.m, and preferably in a range of between about 5 .mu.m and about 30 .mu.m. The thickness of the electrically conducting layer comprising first electrode 22 can also be important. If layer 22 is not recessed into layer 26, then the thickness of layer 22 should be smaller than the thickness of the stratum corneum, i.e., smaller than about 20 .mu.m. Specifically, unless first electrode 22 is recessed into layer 26, the thickness of layer 22 should be in a range of between about 10 nm (10.sup.-8 m) and about 10 .mu.m, preferably in a range of between about 0.1 .mu.m and about 1 .mu.m. The lower bound for the thickness of first

electrode 22 is determined primarily by the objective of having a low electrical resistance and of having a sufficient thickness of electrode material that the electrode is stable with respect to oxidation, electro-erosion and mechanical stress.

Detailed Description Text (8):

Composition 20 is intended to supply or receive transported molecules or ions, and is contained within housing 14 in fluid communication with epidermis 12 through aligned perforations 24 of perforate electrically-insulating layer 26 and first electrode 22. Second electrode 32 is located above perforate electrically-insulating layer 26. Wire 34 connects second electrode 32 to voltage source 36 at switch 38. An example of a suitable voltage source 36 is an exponential voltage pulse generator. In another embodiment, an electric current or charge source can be used instead of a voltage source.

Detailed Description Text (10):

Wire 80 extends from resistance-measurement electrode 31 to terminal 44 of switch 38. Wire 82 extends from terminal 46 of switch 38 to resistance measurement device 84, which includes signal generator 86, amplifier 88, oscilloscope 90 and an electrical ground. Wire 92 extending from first electrode 22 extends to terminal 58 of switch 38.

Detailed Description Text (11):

Electrode 94 is located at epidermis 12 of the patient. Optionally, electrode 94 can be omitted from electrode/reservoir 10. Wire 96 extending from electrode 94 is connected to ground 98. Typically, electrode 94 is located between about five and about twenty centimeters from housing 14.

Detailed Description Text (12):

Filter 39 is located between perforate electrically-insulating layer 26 and second electrode 32. Optionally, filter 39 can be omitted from electrode/reservoir device 10. Filter 39 can be formed of a suitable electrically nonconducting material, such as cellulose, polyimide, etc. In another embodiment of the present invention, filter 39 can be a membrane formed of suitable electrically nonconducting material, such as suitable filters having a pore size in a range of between about 0.2 and about 100 micrometers.

Detailed Description Text (13):

In another embodiment, voids created by perforate electrically-insulating layer 26, aligned perforations 24 of perforate electrically-insulating layer 26, first electrode 22, filter 39, or combinations thereof, can be filled with composition 20, thereby eliminating the need for excess composition 20 in housing 14. Composition 20 should be in fluid communication with epidermis 12.

Detailed Description Text (14):

Optionally, housing 14 can be omitted. This can be accomplished by placing second electrode 32 above perforate electrically-insulating layer 26. Voids created by electrically-insulating layer 20, aligned perforations 24 of perforate electrically-insulating layer 26, first electrode 22, and, optionally, filter 39 are filled with composition 20, thereby providing fluid communication of composition 20 between electrode 22 and epidermis 12.

Detailed Description Text (15):

The electrodes of electrode/reservoir device 10 are typically composed of a suitable conductive material, such as copper, gold, aluminum etc., but can also include semiconductors, such as silicon, germanium, or resistive conductors, such as carbon. Perforate electrically-insulated layer 26 is formed of a suitable electrically nonconducting material, such as a suitable polyimide, Mylar, etc. Housing 14 is typically composed of a suitable material, such as plastics and other non-electrically conductive materials. Generally, the diameter of perforations 24 at electrode 22 are larger than the diameter of perforations 24 at electrically-insulating layer 26, thereby providing an annular gap at the interface between first electrode 22 and electrically-insulating layer 26.

Detailed Description Text (16):

In one embodiment, as can be seen in FIG. 2, at least a portion of the surface of

perforate electrode 22 is provided with a coating of electrically insulating dielectric material 21, thereby providing an electrical capacitance. This capacitance serves the purpose of diminishing or preventing electrolysis at the aqueous solution/electrode interface and thereby reduces gas evolution and bubble formation within the electrode/reservoir device. Such bubbles are generally undesirable, as their presence interferes with the flow of electrical currents along the intended pathways. Suitable materials for an electrically insulating dielectric coating 21 include Teflon.RTM., Mylar.RTM., etc. and oxides such as aluminum oxide and tantalum oxide.

Detailed Description Text (18):

Returning to FIG. 1, the method of the invention includes applying at least one electrical pulse to epidermis 12 by applying a voltage between second electrode 32 and first electrode 22. A localized electric field is formed between second electrode 32 and first electrode 22 that extends within epidermis 12. The amplitude, number, and duration of pulses is sufficient to electroporate epidermis 12 proximate to first electrode 22. Typically, the epidermis is electroporated by applying a voltage from voltage source 36 to second electrode 32 in a range of between about 50 volts and about 500 volts, preferably between about 70 volts and about 200 volts. In one embodiment, the voltage is applied as a series of pulses. In a specific embodiment, the pulse-applied voltage has an average duration in a range of between about 1 milliseconds and about 3 milliseconds per pulse, and the number of pulses employed to cause electroporation of epidermis 12 is in a range of between about 1 and about 10 pulses.

Detailed Description Text (19):

There are two general modes of use for the electrode/reservoir device 10, an electrical pulsing mode or a resistance measurement mode, with the pulse/measurement choice implemented by using switch 38. As shown in FIG. 1, switch 38 is set in the "pulsing mode," such that a voltage pulse produced by pulse generator 36 results in electrical current flow through wire 40, which passes through current sampling resistor 42 and flows to terminal 52 of switch 38, and then (in the position shown) reaches terminal 54 of switch 38, which is connected by wire 34 to conduct the current into electrode/reservoir device 10 to reach second electrode 32, and then passes through electrically conducting fluid (composition) 20, through filter 39, through one or more holes in electrically insulating layer 26, and then into and through epidermis 12, where the current spreads out and reaches perforate electrode 22, with the current exiting electrode/reservoir device 22 along wire 92 to reach terminal 58 of switch 38, and then to terminal 60 of switch 38, which is connected to ground that provides a return path to pulse generator 36. During such pulsing the skin resistance measurement system 84 is connected by wire 82 to terminal 46 of switch 38, and then in the switch position shown, connected to terminal 48, which is connected to ground 74, with this grounding of measurement system 84 serving to protect said measurement system from stray currents and capacitive pickup during pulsing.

Detailed Description Text (20):

During measurements carried out before or after pulsing, two different measurement modes can be used, one without the use of separate electrode 94, and one which involves electrode 94. For the switch position shown for measurement mode selection switch 62 separate electrode 94 is not involved. In this mode, a small resistance measurement current, for example a small amplitude square wave current (as indicated in resistance measurement device 84), flows from square wave generator 86 within resistance-measurement device 84 through wire 82 to terminal 46 of switch 38, then to terminal 44 of switch 38 through wire 80 into electrode/reservoir device 10 to resistance-measurement electrode 31, through electrically conducting fluid (composition) 20, through one or more holes in electrically insulating layer 26, and then into and through epidermis 12, where the current spreads out and reaches perforate electrode 22, then through wire 92 to terminal 58 of switch 38, which is now connected to terminal 56 of switch 38, then through wire 64 to terminal 66 of measurement mode selection switch 62, which in the position shown is connected to terminal 68 of switch 62, and then through wire 72 to ground, thereby completing the measurement circuit to the ground of resistance-measurement device 84. Amplifier 88 and oscilloscope 90 can be used to measure the voltage developed because of the square wave current, which provides a measure of the skin or tissue resistance adjacent perforation electrode 22. Note that, in this resistance-measurement mode, separate

electrode 94 is usually not present.

Detailed Description Text (21):

In the alternative resistance-measurement mode, resistance-measurement selection switch 62 is put into a second position (not shown). With this setting, the resistance-measurement device measurement current cannot flow out of electrode/reservoir device 10 along wire 92 and into switch 62 to reach ground, because switch 62 is connected to terminal 70, which is electrically open (a very high resistance). Thus, resistance-measurement device measurement current instead flows across the epidermis at the site of one or more holes in perforate electrode 22, but in this case more deeply into the epidermal tissue but with significant current spreading, with the current within the epidermal tissue reaching electrode 94 (electrode area 94 is generally much larger than the total area of the holes in perforate electrode 22), and then through wire 96 to ground 98. In this alternative resistance-measurement mode the resistance-measurement current flows through and within the epidermis at two distinct sites: at the electrode/reservoir device 10 and at the separate electrode 94, but only the resistance of the epidermis adjacent electrode/reservoir device 10 is usually altered by pulsing, and this means that the resistance measurement involving the two sites can be used to infer the resistance changes at the site of the electrode/reservoir device 10.

Detailed Description Text (23):

It is also believed that an electric field generated between the first and second electrodes will preferentially be limited to regions at or immediately adjacent to the perforations of the electrically-insulating layer 26 and portions of tissue, such as skin, most proximate to those perforations. Further, electric field lines that extend from one electrode at the tissue to another electrode located away from the tissue will generally limit the depth of the electric field lines within the tissue. Limiting the depth of electric field lines by use of the apparatus of the invention enables electroporation of the surface layer, or stratum corneum, of the skin without stimulating nerve endings, thereby substantially preventing sensation, including pain, and also substantially preventing stimulation of muscles, thereby also substantially preventing associated tissue movement. In other embodiments, the electric field lines can be controlled by, for example, adjusting the amount of applied voltage to the electrodes, to thereby control the amount of sensation.

Detailed Description Text (26):

In another embodiment, the apparatus represented in FIGS. 3 and 4 can be employed. As shown in FIG. 3, tube/reservoir device 100 is located at epidermis 102 of a human patient. Tube/reservoir device 100 includes outer wall 104 and inner electrically-insulating tube wall 106. The alignment of the inner and outer tubes does not need to be concentric. Outer wall 104 defines application opening 108. Inner electrically-insulating tube wall 106 defines application opening 110 and opening 112. A syringe can be fitted to opening 112. Application opening 110 is recessed within outer wall 104. Optionally, application opening 110 does not need to be recessed. Outer wall 104 and inner electrically-insulating tube wall 106 define a first reservoir containing first electrically-conductive composition 118. As shown in FIG. 4, outer wall 104 and inner electrically-insulating tube wall 106 are concentric, but a centric alignment is unnecessary.

Detailed Description Text (27):

Referring back to FIG. 3, inner electrically-insulating tube wall 106 defines a second reservoir containing second electrically-conductive composition 120. First electrically conductive composition 118 is in fluid communication with epidermis 102 at stratum corneum layer 103 and is contained by the first reservoir and by epidermis 102. Second electrically-conductive composition 120 is in fluid communication with epidermis 102 at stratum corneum layer 103 by the second reservoir and epidermis 102. First electrically-conductive composition 118 is partitioned from second electrically-conductive composition 120 by inner electrically-insulating tube wall 106 and by epidermis 102. First electrode 122 is located at the first reservoir and is in communication with first electrically conductive composition 118. Second electrode 124 is located at the second reservoir and is in communication with second electrically-conductive composition 120. Wire 125 extends from switch 128 to second electrode 124. Wire 126 connects terminal 154 of switch 128 through current sampling resistor 146 to voltage source 130. An example of a suitable voltage source 130 is an

exponential voltage pulse generator. An electric current or charge source can also be used instead of a voltage source.

Detailed Description Text (28):

Wire 132 extends from switch 128 to electrical resistance measurement device 136. Electrical resistance measurement device 136 includes signal generator 138, amplifier 140, oscilloscope 142 and electrical ground 144. Electrode 124 can be employed to measure electrical resistance at epidermis 102 by turning switch 128 from connection with voltage source 130 to connection with electrical resistance measurement device 136. A separate electrode 94 (see FIG. 1) can also be used to make an electrical resistance measurement, but in this case the resistance of the skin at a second site, that of electrode 94, is in series with the skin subject to electroporation. Generally the skin resistance at the site of electrode 94 changes insignificantly during pulsing, and therefore this arrangement can also be used to assess skin resistance changes associated with skin electroporation.

Detailed Description Text (30):

First electrode 122 and second electrode 124 of tube/reservoir device 100 are typically composed of a suitable conductive material, such as copper, gold, aluminum etc. Inner electrically-insulated tube 106 is typically formed of a suitable electrically nonconducting material, such as a plastic or other electrically nonconductive material. Outer tube 104 is also typically formed of an electrically nonconductive material, but can also be constructed of a conductive material, such as stainless steel.

Detailed Description Text (31):

Electrically conductive compositions 118, 120 include materials that can be transported, by migration or some other mechanism, across a portion of epidermis that has been electroporated. In one embodiment, either electrically conductive composition 118 or 120 can be a solution, wherein the agent and/or material are solutes of the solution. Examples of suitable solvents include physiological saline, phosphate buffered saline, distilled water with known trace electrolytes, etc. An electrically conductive gel or paste containing a material to be introduced into or through stratum corneum 103 by the method of the invention can also be employed as electrically conductive composition 118 and/or 120. Examples of suitable materials for electrodes 122 and 124 include electrically conductive materials such as aluminum, etc., and also semiconducting materials such as silicon and germanium, and resistive materials such as carbon.

Detailed Description Text (32):

The method of the invention includes applying at least one electrical pulse to epidermis 102 by applying a voltage between second electrode 124 and first electrode 122. Alternatively, a current or charge pulse can be applied. A localized electric field is thereby formed between first composition 118 and second composition 120 that extends around inner electrically-insulating tube wall 106 at application opening 110 and within epidermis 102. The amplitude, number, and duration of pulses is sufficient to electroporate epidermis 102 proximate to first composition 118 and second composition 120. Typically, the epidermis is electroporated by applying a voltage from voltage source 130 in a range of between about 30 volts and about 1000 volts, and preferably between about 70 volts and about 200 volts. In one embodiment, the voltage is applied as a series of pulses. In a specific embodiment, the pulse-applied voltage has an average duration in a range of between about 1 milliseconds and about 3 milliseconds per pulse, but can be as long as 300 milliseconds, and the number of pulses employed to cause electroporation of epidermis 102 is in a range of between about 1 and about 10 pulses.

Detailed Description Text (33):

There are two general modes of use for the electrode/reservoir device 100, an electrical pulsing mode or a resistance measurement mode, with the pulse/measurement choice implemented by using switch 128. As shown in the drawing, switch 128 is set in "pulsing mode," with terminals 150 and 154 connected to each other and terminals 152 and 156 connected to each other. In this mode, a voltage pulse produced by pulse generator 130 results in electrical current flow through wire 126 which passes through current sampling resistor 146 and flows to terminal 154 of switch 128, then to terminal 150 of switch 128, which is connected to electrode 124, and then passes

through electrically conducting fluid (composition) 120, through stratum corneum 103 and epidermis 102, where the current spreads out and reaches electrically conducting fluid (composition) 118, and then to electrode 122 to ground 148. This completes the pulsing circuit.

Detailed Description Text (35):

During measurements carried out before or after pulsing, switch 128 is placed in the other position, so that terminals 154 and 158 are connected to each other, and terminals 156 and 160 are connected to each other. In this mode, a small resistance measurement current, for example a small-amplitude square wave current (as indicated in signal generator 138) flows from resistance measurement system 136 through wire 132 to terminal 156 of switch 128, then to terminal 160 of switch 128, which is connected to electrode 124, and then passes through electrically conducting fluid (composition) 120, through stratum corneum 103 and epidermis 102, where the current spreads out and reaches electrically conducting fluid (composition) 118, and then to electrode 122 to ground 148. This completes the resistance measurement circuit.

Detailed Description Text (37):

It is believed that an electric field generated between first electrically-conductive composition 118 and second electrically-conductive composition 120 will preferentially be limited to penetration of epidermis 102 most proximate to the first and second electrically-conductive compositions. Further, electric field lines that extend from one electrically-conductive composition to another electrically-conductive composition, separated by a distance defined by the thickness, typically about 20 micrometers to about 100 micrometers, of electrically-insulating tube 106, and the parameter $L_{sub.h}$ generally limits the depth of the electric field lines within the tissue. Typically, $L_{sub.h}$ ranges from a value of about zero to about one quarter of the internal diameter of outer wall 104. Controlling the voltage enables control of the depth of electric field lines by use of the apparatus of the invention. Thus, the surface layer, or stratum corneum, of epidermis 102 can be electroporated in a controlled manner without stimulating nerve endings 105, thereby enabling control of sensation, or substantially preventing pain, and also substantially preventing stimulation of muscles, thereby substantially preventing associated tissue movement.

Detailed Description Text (38):

In still another embodiment, the apparatus represented in FIGS. 5 and 6 can be employed, which allows the site(s) of electroporation to be electrically addressed. As shown in FIG. 5, device 200 is located at epidermis 202 of a human patient. Device 200 includes a plurality of first electrodes 204 that contact epidermis 202. Perforate electrically-insulating layer 206 is located at first electrodes 204. Perforate electrically insulating layer 206 also separates multiple first electrodes 204. Conduits 208 are defined by first electrodes 204 and by perforate electrically-insulating layer 206. Housing 210 extends about electrically-insulating layer 206. Composition 212 is contained within housing 210 and is in fluid communication with epidermis through conduits 208. Second electrode 214 is located above perforate electrically-insulating layer 206 and is immersed in composition 212. Wire 216 extends from second electrode 214 to voltage source 218. An example of a suitable voltage source 218 is an exponential voltage pulse generator.

Detailed Description Text (39):

Wires 220, 222 extend from separate first electrodes 204 to switches 224, 226, respectively. Ground wires 228, 230 extend from switches 224, 226, respectively. Wires 232, 240 extend from switches 224, 226, respectively, to microprocessor 242. Switches 224, 226 are separately controllable by microprocessor 242. It is to be understood that additional switches, having the same configuration, are located between microprocessor 242 and other first electrodes of the array shown in FIG. 6. As shown in FIG. 6, first electrodes 204 can be arranged as of an array, whereby electric fields can be formed at individual first electrodes 204, or patterns thereof, to determine a preferred location or locations for electroporation of epidermis 202. Microprocessor 242 controls which of first electrodes 204 can be used to stimulate electroporation. Microprocessor 242 can be employed to control formation of electric fields at individual first electrodes 204 and to determine preferred lateral locations of epidermis for electroporation.

Detailed Description Text (40):

Referring back to FIG. 5, filter or microporous gel layer 244 is located between perforate electrically insulating layer 206 and second electrode 214. Optionally, filter 244 can be omitted from device 200. Filter 244 can be formed of a suitable electrically nonconducting material, such as cellulose, polyimide, etc. Examples of suitable microporous gel materials include agarose, polyacrylamide, etc. In another embodiment of the present invention, filter 244 can be a membrane formed of suitable electrically nonconducting material, such as a filter having a pore size in a range of between about 0.2 and about 100 micrometers.

Detailed Description Text (41):

The electrodes of device 200 are typically composed of a suitable conductive material, such as copper, gold, aluminum, etc., or semiconductors, such as silicon, germanium, or resistive materials, such as carbon. Perforate electrically-insulating layer 206 is formed of a suitable electrically nonconducting material, such as a suitable polyimide, etc.

Detailed Description Text (43):

At least one electrical pulse is applied to epidermis 202 at stratum corneum layer 246 by applying a voltage between second electrode 214 and first electrode 204. Alternatively, a charge pulse or current pulse can be used. A localized electric field is formed between second electrode 214 and first electrode 204 that extends within epidermis 202. The amplitude, number, and duration of pulses is sufficient to electroporate epidermis 202 proximate to first electrode 204. Typically, the epidermis is electroporated by applying a voltage from voltage source 218 in a range of between about 50 volts and about 200 volts. In one embodiment, the voltage is applied as a series of pulses. In a specific embodiment, the pulse-applied voltage has an average duration in a range of between about 1 milliseconds and about 3 milliseconds per pulse, and the number of pulses employed to cause electroporation of epidermis 202 is in a range of between about 1 and about 10 pulses. Microprocessor 242 can be utilized to generate trial voltage pulses. The trial voltage pulses can be used to identify any one or more lateral regions of epidermis 202 with reduced sensitivity. For example, first electrodes can be selectively activated to identify first electrodes most distant from nerve ending 248 of epidermis 202. Once identified, first electrodes 204 that are most distant from nerve endings 248 can then be selectively employed to electroporate epidermis and to deliver composition 212 into epidermis 202 without causing discomfort to, or undesired muscle movement in, the patient. Subsequent pulsing of the affected area allows electroporation without causing discomfort to, or undesired muscle movement in, the patient.

Detailed Description Text (44):

Although it is generally preferred to use the apparatus of this invention with pulses in a range of between about 50 volts and about 500 volts, because such pulses cause electroporation within the stratum corneum, and result in the largest molecular transport, the apparatus can also be used with pulses in a range of between about 5 volts and about 50 volts, because pulses in this range predominantly cause electroporation of the linings of sweat gland ducts.

Detailed Description Text (45):

The apparatus of this invention can also be used to transport molecules across tissue without necessarily causing electroporation. Unlike electroporation of the stratum corneum, which is caused by large pulses (between about 50 volts and about 500 volts at the electrodes) iontophoresis is often caused by application of essentially steady (direct current) relatively small voltages (between about 0.1 volt and about 5 volt) or currents, which transport molecules through pre-existing pathways. In the case of transdermal molecular transport for drug delivery or chemical analyte extraction, the apparatus can be used with iontophoresis by using much smaller voltages, typically in the range of 0.1 volt to 5 volt (see, for example, B. H. Sage, "Iontophoresis" in Percutaneous Penetration Enhancers E. W. Smith and H. I. Maibach, Eds., CRC Press, pp. 351-368, 1995, and references contained therein, and also J. A. Tamada, N. J. V. Bomannon and R. O. Potts "Measurement of Glucose in Diabetic Subjects Using Noninvasive Transdermal Extraction" Nature Medicine 1:1198-1202, 1995). When used with iontophoresis, the skin resistance changes much more slowly, and much less in magnitude, and this skin resistance behavior is believed to be due to changes of ionic composition of solutions with pre-existing aqueous pathways (see, for example, S. M. Dinh, C-W. Luo and B. Berner "Upper and Lower Limits of Human Skin Electrical

Resistance in Iontophoresis" AIChe J. 39:2011-2018, 1993). Thus, the larger skin resistance during iontophoresis means that the electric field is more confined to the stratum corneum than for electroporated skin if iontophoresis is used with the apparatus of this invention, and electrode/reservoir device dimensions should be chosen accordingly. Generally, however, because of the larger skin resistance during iontophoresis, electrode/reservoir devices designed for the use with electroporation serve to also suitably confine the electric field at the lower voltages used with iontophoresis, because of the larger skin resistance associated with iontophoresis. The apparatus of this invention allows transdermal iontophoresis but avoids deeply penetrating electric fields and their associated currents, and this minimizes electrical interactions with the viable epidermis which can cause irritation and other undesired side effects." An additional feature of the apparatus of this invention is that no electric field or currents cross the stratum corneum at sites other than the site of the device, unless a separate electrode 94 is used for combined measurement of the skin resistance at two sites.

Detailed Description Text (49):

The apparatus and process of the invention were partially illustrated by designing, constructing, and using a hand-drilled electrode array version of an electrode/reservoir device with seven (7) holes (350 .mu.m diameter, 300-600 .mu.m apart, with the perforate electrode formed from a 25 .mu.m thick copper layer attached to a 50 .mu.m thick polyimide electrically insulating layer). Several experiments in vitro tested the suitability of the device for transdermal molecular transport. The results demonstrate that charged molecules were transported locally across the skin at the sites of the holes by using gel trapping microscopy (see Pliquett, U. F. et al., "Imaging of Fluorescent Molecule and Small Ion Transport Through Human Stratum Corneum During High-Voltage Pulsing: Localized Transport Regions are Involved," J. Biophys. Chem. 58, 185-204, [1996]).

Detailed Description Text (50):

This experiment involved applying the prototype electrode/reservoir device (a version of FIG. 1) against the stratum corneum side of a human skin preparation mounted on gelled agarose, with the agarose layer serving to inhibit convection. The donor compartment contained PBS+1 mM calcein and the counter (second) electrode. The electrode/reservoir device was pressed to the skin's surface (with about 100 Pa pressure), and a series of $N_{\text{sub.pulse}}=200$ exponential pulses ($\tau_{\text{sub.pulse}} \approx 1$ ms; maximum transdermal voltage $U_{\text{sub.skin},0} \approx 150$ volts (V)) was delivered. The skin was examined by fluorescence microscopy, and calcein (green fluorescence) observed. We observed green fluorescent rings with the same diameter as the holes of the electrode. The center region was considerably dimmer than the edge, probably resulting from the high resistance of the saline compared to the metal electrode. This means that there was not an equipotential across the 350 .mu.m diameter hole, but instead higher fields near the hole edge. Calcein was also found in the gel, but it had diffused laterally within the gel to form a bright region underneath the skin. It nevertheless confirms that transport across the skin took place. Our interpretation is that calcein was successfully transported locally across human skin, as desired.

Detailed Description Text (52):

A very similar electrode/reservoir device was used to demonstrate that very little sensation was perceived in vivo. Specifically, the ability of the prototype device to minimize and/or avoid sensation and pain was tested by two human volunteers (Drs. U. Pliquett and J. C. Weaver). Both human volunteers self-administered a series of progressively larger pulses while holding the prototype array against their forearm. This prototype had 48 holes, each 350 .mu.m in diameter, and was constructed from gold-covered Kapton.RTM. polyimide. Two sites were used: (1) dry skin, and (2) skin hydrated by saline contact for 1/2 hour. Dry skin has much larger $R_{\text{sub.skin}}$, and resulted in less sensation; therefore the more relevant (to transdermal drug delivery) hydrated skin result is briefly described.

Detailed Description Text (53):

The array reservoirs (300 .mu.m diameter holes) were filled with saline, to make contact with the skin. Single exponential pulses ($\tau_{\text{sub.pulse}} = 1$ ms) were self-administered, starting at 10 V. For one subject, (JCW), at approximately 150 V a clear perception (a "faint pricking") occurred, and as the voltage was decreased, the

perception now occurred at smaller voltages. However, throughout, there was no major pain. A few different sites were tried, by moving the entire array (there was no site addressing capability in the prototype). Only slight differences were then experienced. For the second subject, (UP), a voltage of up to 250 V was applied without sensation. Increasing the voltage resulted in a sensation compared to the touching of the hairs at the skin surface and later to a very short prick sensation. Sometimes also something within the vicinity of the electrode was felt.

Detailed Description Text (54):

Ordinarily, the application of "high-voltage" pulses to skin electrodes is intended to stimulate nerves. (Reilly, J. P. (Ed.) "Electrical Stimulation and Electropathology," Cambridge University Press, Cambridge, 1992). It is probably not surprising, therefore, that even a non-optimal "field-confining" electrode/reservoir device with similar HV pulses failed to cause significant nerve stimulation.

Detailed Description Text (56):

The invention was further illustrated by another version of the basic apparatus of FIG. 1 in which the hole diameter was much smaller, viz. $20 \pm 0.5 \mu\text{m}$. These were specified by us for fabrication by a commercial source of laser drilling (Laser Services, Inc., Westford, Mass.), using $25 \mu\text{m}$ thick polyimide pieces with a bonded copper layer of $25 \mu\text{m}$ thickness on one side. The holes were laser drilled starting from the side of the polyimide without copper, to minimize copper deposition onto the hole walls. A center-to-center hole spacing of $300 \mu\text{m}$ was used, with a total of about 1,000 holes. This copper-based electrode system changed with pulsing due to electroerosion, and eventually became unrealistic to use.

Detailed Description Text (57):

In the first experiments with this electrode/reservoir device, a series of single exponential pulses ($\tau_{\text{pulse}} \approx 1 \text{ ms}$) was used with the sterile, saline-filled electrode/reservoir device held firmly against a hydrated site on the forearm of a human subject. An ECG electrode (D102 Ag/AgCl, In Vivo Metric, Healdsburg, Calif.) had been placed about 5 cm away, with electrical connections that allowed either "high-voltage" pulsing or resistance measurements. The resistance was that of a series pathway which crossed the skin at two sites: (1) within the electrode/reservoir device, and (2) the ECG electrodes.

Detailed Description Text (58):

In vivo experiments together with manually recorded resistance measurements were performed initially. For the first subject (JCW), the first sensation ("a faint prick") was felt when the applied voltage (between the electrodes) was 300 V, and at the same time the resistance dropped by about 20%. In another experiment, on a second subject (UP), a sensation was not felt until a voltage of 400 V was applied.

Detailed Description Text (59):

Another in vivo experiment with subject UP involved application of single pulses, starting with an electrode voltage across the electrode/reservoir device of 50 V. Thereafter the electrode pulse voltage was increased in 50 V increments, with both the resistance (in series across the skin at both the electrode/reservoir device site and the ECG site) measured and the subjects' perception of sensation noted. After 250 V, the next pulse voltage was chosen to be 270 V (not 275 V), and a measurable decrease of R_{skin} was observed, but there was no sensation. A further pulse of 300 V also caused no sensation, and still other pulses were applied without any sensation.

Detailed Description Text (60):

The same electrode/reservoir device was then used with subject JCW. The value of the two-skin-site resistance before pulsing was $7.2 \times 10^4 \Omega$, but application of a single 150 V pulse across the electrodes resulted in a sudden drop to $5.2 \times 10^4 \Omega$, without any sensation. The next pulse (170 V) caused a further decrease in resistance to $4.8 \times 10^4 \Omega$, which was accompanied by a slight sensation ("a small prick").

Detailed Description Text (63):

The electrode array of the electrode/reservoir device (FIG. 1) was made from Kapton.RTM. polyimide covered with $15 \mu\text{m}$ of aluminum at one side, which contacted the skin. The array consisted of $50 \mu\text{m}$ diameter holes, with two different spacings

present within the array: 50 .mu.m edge-to-edge and 100 .mu.m edge-to-edge. (This array had been produced from a test pattern for an entirely different application.) The array was mounted on one side of a filter housing (in place of the filter) and the other side of the housing was removed. A counterelectrode (0.2 mm copper wire) was placed between the supporting plastic in the housing. The electrode was mounted on a 10 cc syringe and filled with saline. For resistance measurements, a second electrode (jD102 Ag/AgCl, In Vivo Metric, Healdsburg, Calif.) was placed within the saline and a second one at the skin surface in the immediate vicinity of the electrode/reservoir device. Note that this resistance is the series resistance across two skin sites, with only one altered by localized skin electroporation (FIG. 1).

Detailed Description Text (64):

A "high-voltage" pulse (with .tau..sub.pulse .apprxeq.0.8 ms) was delivered by a custom-built pulser through the electrode/reservoir device. The pulse was applied through a high-voltage switch, automatically disconnecting the impedance measurement system for the duration of the pulse.

Detailed Description Text (65):

The voltage (across the electrodes) of the first pulse was 100 V, and was increased until a sensation was felt. A plot of the resistance vs. electrode voltage for a first subject is shown in FIG. 7, in which the sequence of pulses is labeled numerically as points 1 through 13. Then the voltage was not further increased, but the dynamic resistance behavior investigated at the same or lower voltage (in this case 250 V). In this subject, a hint of sensation occurred at points 3 and 4, with stronger sensation at points 5, 6, and 7. Sensation was no longer felt at Point 8 and beyond. The dynamic changes in resistance were still observed, and are interpreted as evidence for electroporation.

Detailed Description Text (66):

The results of the same type of experiment on a second subject are plotted in FIG. 8, in which the sequence of pulses is also labeled numerically as points 1 through 10. In this subject, no sensation was felt, but the change in the skin resistance strongly support the occurrence of electroporation. The repetition at 200 V did not cause any sensation but did cause reversible changes in the total resistance across the two skin sites (one not pulsed; see FIG. 1).

Detailed Description Text (67):

FIG. 9 shows the time course of the resistance from a single 450 V pulse. A drop in resistance is evident as well as recovery within seconds to about 90% of the pre-pulse value. This too is consistent with electroporation.

Other Reference Publication (5):

Okino et al., "Effects of a High-Voltage Electrical Impulse and an Anticancer Drug on In Vivo Growing Tumors", Jpn. J. Cancer Res. 78(12):1319-1321 (1987).

Other Reference Publication (8):

Heller et al., "Transfer of Human Membrane Surface Components by Incorporating Human Cells into Intact Animal Tissue by Cell Tissue Electrofusion In Vivo", Biochimica et Biophysica Acta 1024:185-188 (1990).

Other Reference Publication (15):

Prausnitz et al., "Methods for in Vivo Tissue Electroporation Using Surface Electrodes", Academic Press, Inc. Drug Delivery 1:125-131 (1993).

Other Reference Publication (20):

Dinh et al., "Upper and Lower Limits of Human Skin Electrical Resistance in Iontophoresis", American Institute of Chemical Engineers Journal 39(12):2011-2018 (1993).

Other Reference Publication (21):

Pliquett et al., "Imaging of Fluorescent Molecules and Small Ion Transport Through Human Stratum Corneum During High-Voltage Pulsing: Localized Transport Regions are Involved", J. Biophysical Chemistry 58:185-204 (1996).

Other Reference Publication (22):

Pliquett et al., "Changes in the Passive Electrical Properties of Human Stratum Corneum Due to Electroporation", Biochemica et Biophysica Acta 1239:111-121 (1995).

CLAIMS:

1. An apparatus for electroporating a tissue, comprising:
 - a) a perforate electrically insulating layer;
 - b) at least one first external electrode contacting a first side of said perforate electrically insulating layer;
 - c) a second electrode having a first surface and an opposite second surface, said second surface being located at a second side of said perforate electrically insulating layer, whereby an electric field extending between said first and second electrodes will preferentially extend through perforations of said electrically insulating layer, said electric field thereby causing electroporation of a tissue that is proximate to said first electrode and is partitioned from said electrically insulating layer and said second electrode by the first electrode; and
 - d) means for conducting a fluid from the first surface of the second electrode to the second surface of said second electrode.
2. The apparatus of claim 1, wherein said first electrode is a plurality of first electrodes, whereby said first electrodes are electrically insulated from each other.
3. The apparatus of claim 1, wherein said first electrode is perforate.
4. The apparatus of claim 3, wherein at least a portion of perforations in said first electrode are substantially aligned with at least a portion of perforations of said electrically insulating layer.
5. The apparatus of claim 4, wherein said first electrode and said perforated electrically insulating layer are in contact with each other at said aligned perforations, whereby said aligned perforations can contain a material to be conducted into electroporated tissue at the first electrode.
6. The apparatus of claim 5, wherein said perforations of the first electrode have a larger diameter than diameter of perforations of the perforate electrically-insulating layer.
7. The apparatus of claim 5, further including a housing for containing a material to be directed into electroporated tissue at the first electrode, said housing defining an application opening at which the first electrode is located, said electrically insulating layer and said second electrode being contained by said housing and said first electrode.
9. The apparatus of claim 8, further including a filter element located between said electrically insulating layer and said second electrode.
10. The apparatus of claim 9, further including an electrical resistance measurement means, comprising:
 - a) an electrical resistance measurement component;
 - b) a first electrical resistance measurement electrode connected to the electrical resistance measurement component and located at said housing; and
 - c) a second electrical resistance measurement electrode for contact with the tissue and connected to said electrical resistance measurement component.
15. The apparatus of claim 10, further including a switching means at said first and second electrodes for switching between application of an electric field and measurement of electrical resistance at the tissue.

16. An apparatus as in claim 1 wherein said first electrode is at least partially coated by a second electrically insulating layer.

17. A method for electroporating a tissue, comprising the steps of:

- a) contacting at least one first electrode to the tissue;
- b) forming an electric field between said first electrode and a second electrode that is partitioned from said first electrode by a nonconductive layer that contacts the first electrode, whereby said electric field preferentially extends around at least a portion of said insulating layer, the electric field thereby causing electroporation of tissue that is proximate to the first electrode; and
- c) forming a plurality of electric fields, each electric field being formed between a first electrode and the second electrode.

18. The method of claim 17 wherein said electric fields are formed sequentially to selectively designate at least one first electrode that is most proximate to a preferred location at said tissue for electroporation.

19. A method for causing iontophoresis in a tissue, comprising the steps of:

- a) contacting a least one first electrode to the tissue;
- b) forming an electric field between said first electrode and a second electrode that is partitioned from said first electrode by a nonconductive layer that contacts the first electrode, whereby said electric field preferentially extends around at least a portion of said insulating layer, the electric field thereby causing iontophoresis of tissue that is proximate to the first electrode; and
- c) directing a material from a first surface of the second electrode to a second surface of the second electrode and into iontophoresed tissue proximate to said first electrode.

20. A method of claim 19 wherein said electric field is formed by applying a voltage between said electrodes in a range of between about 0.1 volts and five volts.

End of Result Set☐ **Generate Collection**

L12: Entry 19 of 19

File: USPT

Jan 7, 1975

DOCUMENT-IDENTIFIER: US 3859195 A
TITLE: APPARATUS FOR ELECTROCHEMICAL PROCESSING

Abstract Text (1):

An apparatus for electrochemical processing characterized by extended surface electrodes maintained at a substantially uniform electrical potential while presenting a relatively low pressure drop to liquids in treatment passed through the apparatus.

Brief Summary Text (2):

Generally, this invention comprises an apparatus for the electrochemical treatment of an electrically conductive solution by effecting separation, reaction or other processing of a given ingredient of the solution. The apparatus is an electrolytic cell comprising at least two inter-functioning electrodes disposed within a vertically oriented leak-tight housing, at least one of which electrodes has an extended surface area over which a substantially uniform reaction-producing electrical potential is maintained throughout that portion of the area of the electrode in confrontation with the electrolyzing area of the remaining electrode, the separatory (or reactive) one of the electrode pair being relatively open to access of electrically conductive solutions passed through the cell, whereas the remaining electrode is isolated from the solution by enclosure within an electrically insulative envelope permitting ionic conduction while barring ready passage of the solution therethrough.

Drawing Description Text (5):

FIG. 3 is a somewhat schematic partially opened view of a preferred embodiment of spiral wrap electrode structure for use with this invention,

Drawing Description Text (6):

FIGS. 4A-4C are partially schematic representations of two different knitted and woven electrode element designs of uniformly reticulated open construction which can be utilized with this invention,

Drawing Description Text (7):

FIG. 5A is a partially schematic side elevation view of a third electrode structure of "cyclone fence" design,

Drawing Description Text (9):

FIG. 5C is a partially schematic fragmentary perspective view of a fourth electrode structure fabricated from expanded metal,

Drawing Description Text (12):

FIG. 8 is a plot of copper removal according to this invention from a typical industrial effluent containing Cu as adulterant wherein Cu concentration is plotted in ppm as ordinate versus time in minutes as abscissa,

Drawing Description Text (13):

FIG. 9 is a partially schematic perspective view of a rectangular electrode embodiment of this invention.

Detailed Description Text (3):

In ordinary electrochemical processing of aqueous solutions, e.g., in electroplating, the ionic components are present in relatively high concentrations and thus migrate to the electrodes at rates consistent with practical current densities and at relatively high coulombic efficiencies for the desired reactions. However, in many situations, such as most aqueous wastes from industrial plant operations, the ionic components are

present at levels two or three orders of magnitude lower than those encountered in plating practice. If electrochemical treatments are to be applied to the low concentrations of contaminants in such aqueous wastes, it is found that conventional apparatus known to the art possesses very low electrical efficiency, coupled with excessive cell residence time, which renders it impractical to reduce the concentration of the contaminants to the safe levels prescribed by law, e.g., one part per million.

Detailed Description Text (5):

Attempts have been made to solve the problems described by utilizing porous carbon electrode equivalents, including fixed and fluidized beds of carbon particles surrounding the metallic bus supplying current, thereby relying only on point-to-point contact of the carbon particles to maintain conduction. Electrodes fabricated from porous carbon have also been used, through the pores of which the liquid electrolyte is forced to flow; however, these suffer from high pressure drop as well as rapid blinding resulting from accumulation of the plated-out component within the fine pores.

Detailed Description Text (6):

Blocks of carbon can be arranged to provide a high area electrode surface per unit volume of the bed; however, there are disadvantages, such as: (1) carbon has a higher specific resistivity than most metals, so that an electron current flowing within a solid carbon matrix displays a substantial voltage drop between the supply bus and the remote regions of electrode surface. Non-uniformity of electrode surface potential permits concomitant multiple electrochemical processes at different electrode regions, instead of restricting operation to a desired single process. (2) Conventional porous carbon has a very fine pore size of, typically, 0.001 inch- 0.006 inch (25 to 150 .mu.) and is, therefore, quickly clogged with an electrochemical deposit. (3) Fluid flow resistance is very high as a result of both fine pore size and low porosity (e.g., typically, the void fraction is 0.5 or less), requiring either high pressure operation or a very low flow rate.

Detailed Description Text (7):

Packed beds of carbon particles can, by careful selection of particle size and shape, afford a more open deposit area for the deposition of electrochemical product, which has a longer service life, before blinding, together with a lower pressure drop. However, a new difficulty is encountered, namely, reliance on point-to-point particles contact for the electrode circuit path, which evinces a higher electrical resistance between the supply bus and the bed extremities, so that current efficiency is reduced even below that of a block carbon electrode structure. Moreover, the void fraction of packed beds is usually only about 0.50, making only half of the bed section available for passage of the solution in treatment.

Detailed Description Text (8):

Fluidized beds of carbon particles display even more severe disadvantages, in that: (1) there is a substantially higher particle-to-particle electrical resistance, because the area of particle contact are smaller and there exist only low interparticle pressure, since the bed is at least partially floated by the process stream throughput, (2) part of the time some particles are completely out of contact with any others, contributing zero electrode activity and, where corrosive solutions are being treated, actually reducing overall efficiency by loss of previously deposited material through corrosive attack, and (3) deposits built up on individual fluidized particles change the fluidization properties of the particles, making for objectionable inconstancy and nonuniformity of electrode behavior.

Detailed Description Text (9):

Use of metallic extended surface electrode structures is disclosed in U.S. Pat. No. 2,588,450 in the form of a loose stainless steel wool pad as cathode disposed within a cylindrical basket fabricated from an insulative material which is perforated on the periphery to divert the electrolyte flow to an anode disposed coaxially with respect to the cathode. The cell is utilized for the electrodeposition recovery of gold from caustic solution, and the electrolyte is introduced first into the center of the cathode, after which it flows generally laterally through the basket side wall and thence to the encircling anode, so that contact of electrolyte with the electrodes is sequential from cathode to anode.

Detailed Description Text (10):

U.S. Pat. No. 3,244,604 shows electrolyte flow-through woven mesh cathodes 1/4 to 4 inches thick, used for removing metal ion impurities which are present at concentrations below about 500 ppm in aqueous caustic solutions. The cathodes are pads of woven nickel wire stiffened by a nickel screen to which electrical connection is made. Anodes and cathodes alternate in vertical array in the direction of electrolyte flow and there is, of course, no separator inhibiting electrolyte flow-through with respect to the anodes, since the electrolyte has to flow with equal facility through all of the electrodes in the vertically disposed stack.

Detailed Description Text (14):

1. The equilibrium reversible half-cell potential at the conditions of the electrolysis (i.e., temperature, pressure, concentrations of species at the electrode-electrolyte interface, etc.),

Detailed Description Text (15):

2. The activation overpotential, this being the extra electrical potential over and above the equilibrium reversible half-cell potential required to drive the desired reaction at a given rate. This overpotential is a function of the real current density at the electrode-electrolyte interface, and

Detailed Description Text (16):

3. Ohmic voltage drop in the electrode material. This voltage drop is the integral sum of the ohmic voltage drops along each of the paths of current flow leading from the electrode-electrolyte interfaces to the said point in the electrode.

Detailed Description Text (17):

This invention advantageously influences all three of the factors determinative of electrode point potential in the following respects:

Detailed Description Text (18):

a. The electrodes, constructed of extremely fine filaments which provide large surface area and enhance the mass transfer coefficient, minimize the concentration difference between the bulk of the electrolyte and the electrode-electrolyte interface at any given level of operation. Correspondingly, the departure from the equilibrium reversible half-cell potential predicted from the bulk concentrations is minimized.

Detailed Description Text (20):

b. The activation overpotential, which is a function of the current density, is minimized at any given level of operation by the high real surface area of the extended surface electrode. This contributes even more importantly than (a) supra to selectivity and enhanced coulombic efficiency for the desired reaction.

Detailed Description Text (21):

c. The high conductivity of the preferred electrode materials, together with the use of welded or other low resistance connections to buses, minimizes ohmic voltage drop for any given level of operation.

Detailed Description Text (22):

By reduction of each of the enumerated voltage contributions, the power required for a given level of cell operation is substantially reduced. A further contribution to higher power efficiency is the close juxtaposition of anode to cathode in cells of this invention. This is achieved by enclosing one of the interfunctioning electrodes of the pair within a separator presenting both sides of this isolated electrode in electrolyzing disposition with respect to a co-functioning electrode.

Detailed Description Text (25):

The cell of FIG. 1 is provided with two concentric cathode element sleeves 14 and 15 coaxially disposed with respect to the common axis of enclosure 11 and anode 10, although a greater or lesser number can be utilized if desired. An essential feature of this invention is the utilization of cathode elements which have a very large extended surface, while at the same time insuring substantially equipotential maintenance over the electrochemically effective surface as well as presenting an open structure permitting low pressure drop passage of electrolyte therethrough. Bulky open

mesh designs of the structures shown in FIGS. 4A and 4B have proved highly effective because of their uniformity in construction and because of their physical flexibility in assembling apparatus such as those shown in FIGS. 1 and 3. However, uniformly reticulated structures generally, such as the species shown in FIGS. 4C and 5A-5C, can also be used.

Detailed Description Text (26):

Extended surface electrodes employed in my cells can be made from evenly distributed highly conductive continuous run corrosion-resistant metal filaments 2 to 4.5 mil dia., fabricated into knitted form having a void volume in excess of about 85 percent and a surface area in excess of about 5 cm.²/cm.³ of electrode volume. While FIG. 1 details extended surface cathodes, it will be understood that, where anodic electrodes are to be utilized in electrochemical processing, the same electrode structures are ideal for anodic service and, in fact, the electrical supply leads can be simply reversed in polarity to convert cathodic operation into anodic operation. Cathode bus connections are effected through branched upright bus connectors 17a, weld attached to radially opposed pairs of cathodic elements 14, 15 and attached at the bottom ends to a cathodic bus 17 extending radially out through the wall of enclosure 11 to a suitable conventional d-c source, not detailed.

Detailed Description Text (27):

As shown in FIG. 1, anode 10 is mounted within a spacer cage 20, which can be of generally open cylindrical form constituting circular strips 20a joined on the inside surfaces to upright strips 20b, pairs of which latter elements define vertical passages between them for escape of gas released at anode 10. A porous construction is preferred for spacer cage 20, a preferred material of construction for an electrically insulating cage being a unitary bulky polymeric product (e.g., polyethylene) produced by extrusion through a rotating grooved annular extrusion die. The product is an open net-like structure made up of coarse filaments laid over one another at right angles and bonded at crossing points.

Detailed Description Text (28):

Optionally, an electrically conductive (metal) spacer cage of generally similar construction as that hereinabove described can be employed, in which case cage 20 constitutes a radial extension of anode 10 since it is in conductive contact therewith.

Detailed Description Text (29):

Enclosing the anode-spacer cage subassembly is a porous cup-like nonconducting electrode separator 23 having its base end 23a disposed in the direction of process liquid input which, in FIG. 1, is shown as from the bottom as denoted by the directional arrows. Separator 23 can be typically fabricated from spunbonded polyethylene.

Detailed Description Text (30):

The purpose of separator 23 is to forestall electronic conduction between anode 10 and cathode 14, 15 by isolating against metal-to-metal electrode contact, while still furnishing an ionic passage through the solution spanning the electrodes. The permeability of separator 23 should be moderately high for good ionic conductivity while still being low enough to prevent gross mixing between the anolyte and catholyte solutions.

Detailed Description Text (31):

It is also permissible to fabricate separator 23 from electrically insulative ion-exchange membrane materials, such that ions of at least one component in the solution can pass through the separator, thereby completing the ionic current path, while forestalling direct metal-to-metal electronic conduction between the electrodes within the cell as well as direct solution flow between electrolyte chambers.

Detailed Description Text (32):

In summary, the cell structure described supra utilizes very large extended surface electrodes arranged so as to (1) accommodate liquid electrolyte flow therepast with only moderate pressure drop, (2) provide short path distance from any point in the electrolyte in process to the effective extended surface electrode in order to facilitate rapid ionic discharge and (3) insure exceedingly uniform potential

maintenance between bus connectors 17a and cathodes 14, 15 by keeping the electrical resistivity to a minimum not only within the electrode structure per se but also from point-to-point throughout the electrode structure en route to buses 17.

Detailed Description Text (33):

The absolute magnitudes and relative quantitative importance of the physical parameters of the extended surface electrodes described depend, importantly, upon the particular electrochemical process to be effected. For example:

Detailed Description Text (34):

1. In the cathodic deposition of heavy metals such as Cu existing in low concentrations in dilute H₂SO₄ solutions, the current densities required for stoichiometric equivalence to the metal content removed, with conventional liquid flow velocities through the cathode, will be relatively low. The voltage drop across the conductive path between supply bus 17, bus connectors 17a and the extremity of any filamentary path in the cathode mesh is the product of current (I) and path resistance (R). If fine filaments are used in the cathode structure in order to secure large surface per unit volume of the mesh, the increase in R resulting from the small diameter filaments is largely offset by the high conductivity. Thus, the existing IR drop is relatively minimal, so that the voltages at all points on the cathode surface are approximately the same as the supply voltage of bus 17 and bus connectors 17a.

Detailed Description Text (35):

2. In the cathode removal of one metal while preserving an accompanying metal of nearly equal deposition voltage in solution, it becomes far more important that in (1) supra to assure a close approach to equipotential cathode surfaces, and filaments of heavier gage are preferred in this regard.

Detailed Description Text (36):

3. When the concentrations of materials being treated are higher than in the usual wastes, e.g., in electrowinning, the operation is less affected by rate of mass transfer of ions to an electrode because more ions are available and the current densities are higher in meeting the stoichiometric requirements, so that it becomes even more important that in (1) and (2) supra to assure equipotential electrode surfaces. Here it is often feasible to sacrifice some electrode open fluid space, concomitantly increasing the conductive filament cross-sections.

Detailed Description Text (37):

4. As an extension of case (3) supra, in situations where no deposit forms on the electrodes, e.g., anodic oxidation or cathodic reduction forming soluble products, likelihood of plugging porous electrodes is greatly reduced, therefore it is practicable to substitute lower porosities in favor of heavier electrode cross-sections.

Detailed Description Text (38):

Referring now to FIG. 3, there is shown a preferred design of cell according to this invention employing one-piece spirally wrapped cathode, anode electrode structures which are particularly advantageous in providing a large electrode surface in form adapted to insertion into a tubular, leak-tight housing, such as a conduit. Elements in FIG. 3 corresponding to the same elements in FIG. 1 are denoted by the same reference numerals, except that these are primed. The external housing 11 is omitted in the showing of FIG. 3 and, to more clearly expose the anode-spacer structure, the outer terminal portions of the interwrapped elements are pulled apart radially to bring sizable surface portion into better view.

Detailed Description Text (39):

The cathode, or outer member shown in FIG. 3 is portrayed as having three schematically represented cathode elements, i.e., 14', 15', and 25, each consisting of a flattened stainless steel wire tubular knit structure pressed into tight electrically conductive contact with its neighbor. Also, since anode 10', which can be a screen or foil of electrochemically inert material (e.g., platinum), is effective on both of its side surfaces, individual spacers 20' and 20" are employed on opposite sides of the anode, the pair constituting the equivalent of spacer cage 20, FIG. 1. The equivalent of cup-like separator 23 is the sharply folded double ply element 23'bottom, which encases the subassembly 20', 10', 20" on the bottom edge and both

sides, it being understood that the process liquid again is supplied from the bottom, as denoted by the directional arrow, whereas exit of treated liquid and any gaseous products is via the top.

Detailed Description Text (40):

Positive supply bus branches 12', 12' are advantageously spaced along the length of anode 10', to better distribute the supply of electrolyzing current and voltage in accordance with the demand imposed by equal incremental lengths of anode surfaces. The same principle is applied with respect to negative supply bus connectors 17a', 17a'. The structure of FIG. 3 is shown in somewhat loosely rolled condition, so that there exists a longitudinal passage 26 at the inner extremity of the spiral. With tighter roll wrapping, this bore virtually disappears, or can be plugged by a non-conducting rod, so that no bypass through the cell is presented to process solution flowing axially thereof.

Detailed Description Text (41):

The structure of FIG. 3 can be neatly fitted into a circular tube enclosure and, even though it is tightly wrapped into very compact space, only moderate resistance is interposed to solution flow.

Detailed Description Text (42):

Referring now to FIGS. 4 and 5, there are shown details of extended surface electrode formation. Thus, FIG. 4A (and end view 4B) show a knitted bulky wire structure, such as used in the apparatus of FIG. 1, made by interlooping adjacent courses of long length wire denoted 27, which are weld-joined to bus connectors 17a.

Detailed Description Text (43):

Very satisfactory operation has been obtained using cathode mesh structures of the configuration shown in FIGS. 4A and 4B employing from about twenty to about one hundred and twenty individual wire layers. It is a convenience to utilize flattened tubular knit sleeves fabricated from Type 316 stainless steel wire having two filaments in each loop, the filament diameters being any of 2 mil, 3 mil or 4.5 mil (corresponding, respectively, to 0.051 mm, 0.076 mm and 0.115 mm diameters), as marketed commercially by Metex Corporation, Edison, N.J. Referring to FIG. 3, and utilizing a single spacer made up of Du Pont Vexar polymeric netting measuring about 15 mils (0.38 mm) thick and a separator 23' measuring about 6 mils (0.152 mm) thick, it was practicable to obtain a four-wrap cathode structure, each wrap of which constituted twenty-five overlaid double layer flattened knit sleeves, or a total of two hundred radially disposed individual mesh layers, which assembly fit snugly into a Pyrex pipe enclosure having an inside diameter of 51 mm.

Detailed Description Text (44):

A great variety of materials having a wide range of dimensions can be utilized in extended surface electrodes as taught in this invention, however, the following general combination has given exceptionally good results: (1) spiral arrangement of electrodes as shown in FIG. 3, (2) use of a pair of spacers 20' (one on each side of the anode) insuring unhindered egress of any gases generated, especially at the anode 10', (3) a separator constituting an envelope open only along the cell electrolyte outlet side made up of a strong web material such as ion exchange membranes, textile fabrics, paper and spunbonded products having a permeability to ionic passage equivalent to a pore size of about 0.2 mil, (4) knitted mesh of continuous metallic filaments as basis for the extended surface electrode such as shown in FIGS. 4A and 4B, (5) electrode mesh size 6-20 courses/inch (2.4-8 courses/cm), (6) extended surface electrode area 30-50 cm.²/cm.³, (7) ratio of electrode area cathode/anode 5-50 in service of removing trace heavy metals from waste solutions, (8) void volume in extended surface electrode, 90-95 percent, (9) extended surface electrode filament size, 2-5 mils (50 .mu.), (10) treated solution flow velocity, 1-10 cm/sec, and (11) anode current density, 5-200 ma/cm.², based on the anode's simple projected area (i.e., the product of width x height).

Detailed Description Text (45):

Cells constructed and operated as described supra have the following characteristics: (1) average distance from any point in liquid in process flowing through extended surface electrode to a filament surface is 0.7 mm., (2) when operated at a high anode current level (e.g., 60 ma/cm.²) the maximum voltage drop from the bus bar to the

most remote location in the extended surface electrode structure is less than about 0.160 volt and (3) the pressure drop through a typical 51 mm. dia. "jellyroll" spiral electrode assembly for a 75 mm. active length of the roll of extended surface electrode (cathode) is as follows for two sizes of filament in knitted mesh (FIG. 4A design):

Detailed Description Text (47):

In this structure the loops are woven in as part of the warp and constitute the wire structure of the electrode. Filler fibers 32 run at right angles to the loops and can, or need not, be electrically conductive. The purpose of the filler fibers is primarily to complete the weave and hold the entire structure together for mechanical integrity. If filler fibers 32 are conductive, it is preferred that their ends be joined to an additional bus structure in a manner as hereinbefore described for filaments 30 and 31 in order to insure lowest possible electrical resistance within the cathode structure materials. Warp filament 33, and others not shown but reversed in orientation with respect to opposite individual loops, are companionate threads binding the filler fibers 32 in place. If the warp filament 33 is conductive, it is preferably welded at the end to bus connector 17a. However, it need not be conductive and, in this case, circuit contact with the bus is dispensed with.

Detailed Description Text (49):

FIG. 5A, a plan fragmentary view, and FIG. 5B, an end view, together show a "cyclone-fence" type electrode structure made up of helically interwound adjacent metal filaments, such as 39a, 39b, weld-joined at their ends to cathode bus connector 17a. The relatively large diameters of the helices (refer FIG. 5B) constitute, in multi-layer assemblages, an exceptionally open electrode structure not readily plugged by suspended solids in the electrolyte in process.

Detailed Description Text (50):

Finally, FIG. 5C shows, in perspective, a conventional expanded metal electrode structure in which the sheet 40 is first provided with a line of slits and thereafter pulled longitudinally to open the slits into diamond apertures 41. The outwardly projecting ends are weld-joined to cathode 17a and successive sheets 40 are creased or corrugated transversely along lines 42 to furnish the desired separatory offset between adjacent sheet members.

Detailed Description Text (51):

The foregoing description is particularly directed to the preferred embodiment "jelly-roll", or spiral assembly, cell construction; however, extended surface flow-through electrodes having high conductivity and porosity can equally well be assembled in flat stack form, resembling a "club sandwich" arrangement. Thus, this invention is readily applicable to commercial plate-and-frame electrolysis cell designs and the like.

Detailed Description Text (52):

Referring to FIG. 9, there is shown a cell embodiment according to this invention utilizing planar electrodes which, in this instance, are disposed in vertical coparallel rectangular array with the solution to be subjected to electrochemical processing introduced from the bottom, as indicated by the flow directional arrow. The containment housing is omitted in this FIG. for simplicity in the showing as is also the right-hand cathode sub-assembly, it being preferred to associate each anode with a pair of cathode assemblies, one on each side.

Detailed Description Text (53):

The anode 70 is, in this instance, a solid metal plate, typically fabricated from platinum about 25 mils thick. Anode 70 is disposed within a separator envelope 71 of good ionic permeability, as hereinbefore described, which nevertheless effectively isolates the anode from ready circulation of electrolyte liquid through the anode region. Spacers 72, also constructed as hereinbefore described, are interposed between each face of anode 70 and the inside faces of separator envelope 71.

Detailed Description Text (57):

For purposes of simplification of the following description, it is assumed that unit 47 is a single cell. Then, after a given period of operation, depending on the size of the cell of unit 47 and the quantity of metal which is removed from the waste

solution, the cathode space of the cell becomes partially filled with the deposit of the metal which plates out upon the elements of the cathode structure. This increases the pressure differential across the cell to a point where it is economical to remove the cell from service briefly for regeneration. This is conveniently effected by the balance of apparatus shown in FIG. 2.

Detailed Description Text (58):

Thus, a moderately concentrated acid solution, such as nitric acid, is stored in leachate tank 51 so that, when electrolysis is halted in unit 47, the acid is forced, by pump 52, through the electrolytic cell of unit 47 with return back to tank 51. The leaching solution speedily dissolves plated-out metal from the cathode structure to produce a highly concentrated acid leachate solution thereof. The leachate solution can be accumulated in tank 51 until a convenient time arrives to subject it to electrolytic recovery in a conventional electrolytic recovery unit, denoted generally at 54, to which the solution is supplied via pump 53. The acid, stripped of its metal content, is recycled back to tank 51 via line 56 for repeated regeneration service. The conventional direct current power supply, denoted generally at 58, is shown as furnishing electrolyzing current independently to both units 47 and 54.

Detailed Description Text (59):

Others methods exist for removing accumulated metal from the extended surface electrode of unit 47. Thus, a plugged cell can be taken out of service and a fresh cathode substituted for the filled cathode structure, after which operation is restored. Then, in a separate system, the clogged electrode is made anodic with respect to another electrode immersed in a small volume of concentrated electrolyte, and metal is anodically dissolved away to produce a highly concentrated metal solution resembling the leachate solution.

Detailed Description Text (60):

It should be mentioned that the electrolytic cells of this invention are well-suited to multiple use and, in this connection, can be readily employed in either series, parallel or combination series-parallel liquid flow convention as dictated by the circumstances. Thus, in one instance where the waste stream involved has a very large volumetric flow rate, a multiplicity of small-sized cells were employed in parallel connection in preference to designing a single large cell to carry the entire load. Similarly, in cases where a considerable quantity of metal is to be removed from a waste solution, it can be convenient to treat the waste stream by series flow through a succession of extended surface electrode cells, the first of which, in order, remove the large quantities of the metal whereas later ones in the series clean up the traces.

Detailed Description Text (62):

The electrical supply for the hereinabove described cell arrangements is completely independent of the liquid flow conventions. Thus, if multiple cells are operated in liquid flow parallel, all of the potentials required for equal cell plateout would be the same, so that electrical parallel power supply is advantageous. Conversely, if the cells are arranged in liquid flow series, each separate stage usually has a particular optimal operating voltage, in which case separate power supplies of preselected voltage output are usually preferred.

Detailed Description Text (67):

A cell was assembled as detailed in FIG. 3 incorporating an extended surface cathode, a polymeric separator, two polymeric spacers and a screen anode layered together as a sandwich-like stack which was then rolled into a spiral for insertion in tubular methyl methacrylate polymer tube.

Detailed Description Text (68):

For this test the cathode (14', 15' and 25') was knitted sleeve material 0.002 inches (50.8 microns), 2-filament, S.A.E. 316 stainless steel mesh having ten courses/in (3.9/cm). The separator 23' was Du Pont Tyvek 1058, a spun-bonded high-density polyethylene of 1.6 oz/yd.^{sup.2} (0.0054 gm/cm.^{sup.2}), about 6 mils (152 .mu.) thick, with individual fibers in the range of 0.2 mil (5 .mu.) dia. The spacers 20' and 20" were Du Pont Vexar 30 CDS 89, an extruded netting of high density polyethethylene, having a thickness of 30 mils (0.76 mm) with eight strands/in. crossing each other at a 90.degree. angle, giving a diamond pattern. The anode 10' was 80 mesh (31.5 mesh/cm)

woven platinum screen having individual filaments 0.0042 inch (107 .mu.) dia.

Detailed Description Text (69):

The cathode was made up of sixty folds of sleeve 2.5 inches (6.35 cm) wide (i.e., four pieces, each folded 15 times constituting 120 individual layers), weighing a total of 60 gms.

Detailed Description Text (70):

The anode dimensions were 2.5 (6.35 cm) wide .times. 6 inches (15.2 cm) long. The two Vexar spacers were the same length as the anode, but 2.75 inches (7.0 cm) wide. A piece of Tyvek 7 (17.8 cm) long .times. 6.0 inches (15.2 cm) wide constituted the separator. It was folded around the spacers and the anodes and then placed on the layered cathode. The electrodes and interleaved components were then rolled into a tight spiral and inserted into the 2.0 inches (5.08 cm) inside diameter methyl methacrylate polymer tube.

Detailed Description Text (72):

Operation was conducted pursuant to mode 1 (FIG. 6A) except that, after each analysis, the treated solution was returned to vessel 60 for another pass through the cell 47'. This cycle was repeated four times.

Detailed Description Text (75):

The solution velocity (superficial) was 100 cm/min, giving a residence time of 4.2 secs/pass.

Detailed Description Text (82):

The separator was Tyvek 1058 and the spacer was Vexar PDS 89, a polypropylene netting 15 mils (380.mu.) thick. Due to space limitations, only one layer of Vexar spacer was used with the anode in the Tyvek separator envelope. As in EXample I the anode was a 2.5 .times. 6.0 inch piece of 80 mesh woven platinum screen. The components were layered into a sandwich, rolled into a tight spiral and inserted into a glass tube 1.0 inch in dia.

Detailed Description Text (90):

A cell was constructed as described for Example III, except for the following: SAE 304 stainless steel was used as cathode, no spacers were included in the separator envelope and the anode was 80 mesh platinum screen measuring 2 (5.1 cm) .times. 4 inches (10.2 cm).

Detailed Description Text (91):

The solution treated was a sample of an actual industrial effluent "A". It had previously been determined that the composition of this waste stream fluctuated over a wide range, depending upon the particular plant operations in progress at any given time. However, the sample tested here had an initial soluble copper content of 15.5 ppm, a pH = 2.8 and a chloride content of approximately 900 ppm. There were traces of other heavy metals, e.g., Fe, Ni and Cr, and a heterogeneity of dissolved organics, each in low concentrations. Sulfate was the principal anion present.

Detailed Description Text (94):

The cell of Example V was operated in the same manner as described for Example V but used to treat a sample of a different actual industrial plant effluent "B". This aqueous effluent was variable and heterogeneous in composition, but, as received for test, contained 48.5 ppm of copper, an approximately equal content of iron and had a pH = 1.5 due largely to the presence of sulfuric acid.

Detailed Description Text (102):

The cell of Example III was operated in mode 2 (FIG. 6B), that is, the solution treated was circulated from solution accumulation vessel 60' through cell 47", and thence back to vessel 60', which latter was well-stirred during the test. Samples of solution were withdrawn via stopcock 63' for analysis at preselected time intervals denoted.

Detailed Description Text (114):

A cell was constructed without an extended surface cathode to demonstrate the comparative advantages of such electrodes.

Detailed Description Text (115):

Thus, two platinum foil electrodes, each 5 mils (127 microns) thick with dimensions of 2 cm .times. 5 cm, were positioned longitudinally 1.2 cm apart and facing each other in a 1 inch dia. glass tube. The liquid to be treated was circulated through the tube and past both electrodes.

Detailed Description Text (118):

To demonstrate a process application of extended surface electrodes, the system shown in FIG. 2 was assembled.

Detailed Description Text (120):

The aqueous solution treated was an acidified dilute solution of copper sulfate containing 20 ppm of copper at a pH = 2.5. Sulfuric acid was used for pH adjustment. The solution resistivity was adjusted to 40 ohm-centimeters by the addition of sodium sulfate. This solution was pumped through the ESE unit at a rate of 3 gpm (11.7 liters/min), giving a superficial fluid velocity of 9.7 cm/sec. Thus, for the five cells in series, each cell having a working length in the flow direction of about 7 cm, the contact time for the solution treated was approximately 3.6 seconds.

Detailed Description Text (121):

Each cell was operated at constant current by its individual power supply. The applied voltages were low; for example, throughout the 14 hours of the run, the voltage to the first cell never exceeded 5.25 volts, even though the current 17 amps. Because lower currents were applied to the other cells, their voltages were correspondingly lower.

Detailed Description Text (122):

The effluent copper concentration was measured at intervals for both the feed solution and for the solution exiting from each of the five cells. The results are set out in appended Table 13, along with the pressure drop (.DELTA.P) across the cells. Thus, the column headed "Cell I" reports the copper concentration, in ppm, of the solution exiting the first cell, the column under "II" gives the same information for fluid exiting the second cell, etc. The pressure drop across the first cell is .DELTA.P.sub.1, whereas the total pressure drop across all five cells is .DELTA.P.sub.5. Of course, the pressure drops increase with passage of time due to the gradual accumulation of copper in the cathode structures.

Detailed Description Text (123):

At this point in the operation of the system, the input of fresh solution (20 ppm of Cu content) was halted and the power supplied switched off. The column of cells was drained, flushed with water for 30 seconds, drained again, then contacted with an acidic leaching solution to remove the accumulated copper from the stainless steel cathodes.

Detailed Description Text (124):

For the leach operation, a volume of 9.0 liters of approximately 20 percent nitric acid was used. It had been used once previously for a similar leaching operation, so that its copper content was 22,800 ppm (2.28 percent) as initially fed to the cell column. The leachant was circulated through the cell group assembly at a rate of approximately 6 liters/min for a period of 24 minutes. At that point, no further traces of copper were seen to remain in the cell column and leaching was discontinued, the column was drained, then flushed with water for 30 seconds and drained again.

Detailed Description Text (126):

The cycle hereinabove described was thus 14 hours of extended surface electrode operation; drain, leach, etc., approximately 30 minutes.

Detailed Description Text (128):

Recovery unit 54 was a conventional electrolytic copper recovery cell housed in a methyl methacrylate polymer tank 15.2 cm wide .times. 15.2 cm deep .times. 36.8 cm long. The leachate was pumped into the tank at one end near the bottom and flowed over a weir 12.7 cm above the tank bottom before exiting at the opposite end of the tank. A multiplicity of alternated flat anode and cathode plates constituted the electrodes of the recovery cell, these being hung transverse the tank from polymeric holders, so that the plates were at 90.degree. to the general direction of solution flow. Two

cathode plates were interposed between three anode plates, all electrode plates measuring 10.2 cm wide .times. 12.7 cm high, separated one from another a distance of 1.9 cm and immersed to a depth of 10.2 cm. The anodes were 80 mesh (31.5 mesh/cm) platinum screens made up from filaments 107 um dia. The cathodes were 16 gauge type 316 stainless steel plates.

Detailed Description Text (130):

Power was supplied to the electrode of recovery unit 54 from a separate d-c supply at a rate of 22 amperes. This corresponds to a cathodic current density of 50 ma/cm.^{sup.2}. After 5 hours of operation, a total of 99 gms of copper had been deposited on the two cathodes. The copper concentration of the leachate had been reduced to 29,800 ppm. Overall, the electrical efficiency for recovery of copper from the leachate was 76 percent.

Detailed Description Text (134):

For this test an extended surface cathode cell was assembled as follows: the anode was 80 mesh (31.5 mesh/cm) woven platinum screen made up from individual filaments 0.0042 inch (107 .mu.m) dia. The anode was a single sheet measuring 2.5 (6.4 cm) .times. 5.0 inch (12.7 cm). The cathode was 15 layers of knitted 0.002 inch (50.8 .mu.m) 2-filament, type 316 stainless steel mesh 2.5 (6.35 cm) .times. 5.0 inch (12.7 cm), weighing 14.3 gms. A single piece of Vexar screen, of the type described for Example III, was placed beside the described anode into a Tyvek envelope, of the type described in Example III, the whole subassembly being placed in a 60 mm dia. whatman extraction thimble cut off at the closed end to form an open cylinder. The whatman extraction filter is marketed by w. & R. Balston, Ltd., Maidstone, Kent, England, and has the shape of a hollow cylinder closed at one extremity by a hemispherical end. It is made from heavy paper pulp, having a thickness of about one mm, and is used in standard laboratory extractions by the Soxhlet technique. The cell separator layer was cut to fit smoothly once around the cylindrical cathode with no overlap at the ends. The Whatman filter served as a peripheral seal against electrolyte bypassing around the electrolyzing apparatus. The entire assembly was placed inside a 1.0 inch (2.54 cm) inside diameter glass tube so as to give the configuration shown in FIG. 3.

Detailed Description Text (135):

To start this run, 400 cc of effluent "G" solution was placed in a reservoir vessel 60', FIG. 6B, and recirculated (mode 2) through the cell hereinabove described at a rate of 300 cc/min (corresponding to a superficial linear velocity of 1.0 cm/sec). Current to the cell was then supplied at 500 ma, which required approximately 10 volts across the cell. Every hour, 5-10 cc samples of the solution in the reservoir 60' were taken for spectrophotometric analysis by measuring the absorption peak at 525 m.mu.. During the course of this experiment, the destruction of the red-colored dye was seen to occur visually. The data taken were as follows:

Detailed Description Text (139):

$2 \text{ CN}^{\text{sup.}} + 4 \text{ OH}^{\text{sup.}} = 2 \text{ CNO}^{\text{sup.}} + 2 \text{ H}^{\text{sub.2}} \text{ O} + 4 \text{ e}^{\text{sup.}}$, which reaction has a potential of 30 0.970 v. vs. the standard hydrogen electrode (refer "Standard Aqueous Electrode Potentials and Temperature Coefficients at 25.degree.C." by A. J. de Bethune and N.A.S. Loud).

Detailed Description Text (141):

The extended surface area was fabricated as follows: The extended surface area anode was 80 mesh (31.5 mesh/cm) woven platinum screen fabricated from filaments 0.0042 inch (107 .mu.m) dia. Five layers of this screen, each formed by folding a 12 inch long piece back on itself to form a 6 inch length, were used for the anode, forming an assembly 2.5 (6.35 cm) .times. 6 inch (15.2 cm) and weighing 59.5 gms. The cathode was made from a single piece of the same platinum screen measuring 2.5 (6.35 cm) .times. 6 inch (15.2 cm). A single piece of Vexar screen of the type described in Exampale III was placed, as a spacer, along with the described cathode, into a Tyvek envelope separator of the type described in Example III. The whole subassembly was placed in a Whatman extraction thimble of the type described in Example XV and cut to the dimensions of the subassembly in the manner hereinbefore described in Example XV. The entire assembly was placed inside a 1.0 inch (2.54 cm) inside diameter glass tube to give a configuration such as shown in FIG. 3.

Detailed Description Text (143):

A second experiment was made under the same conditions on a solution prepared exactly as described for Experiment No. 1 supra, but containing only 103 ppm CN.sup.- ion.

Detailed Description Text (149):

The cell of Example XVI, with the extraction thimble wrapping replaced by a single wrap of Teflon fluorocarbon sheet measuring 0.031 inch (795 .mu.m) thick and 2.5 inch (6.4 cm) wide, was used to reduce an aqueous quinone solution of approximately 100 ppm concentration to hydroquinone by making the extended surface electrode the cathode.

Detailed Description Text (157):

This Example demonstrates the necessity for spacers 20, 20' and 20", as shown in FIGS. 1 and 3, respectively.

Detailed Description Text (159):

The aqueous solution treated was an actual industrial plant waste containing a varying amount of copper ions in the range of 1 to 10 ppm. The solution was filtered and adjusted to a pH of usually about 3 before it was passed to the cells. Each cell was operated at a current density appropriate to the copper level in the solution at the point of cell entrance, and the current supply ranged from about 2 to about 8 amperes/cell, giving current densities from about 10-40 ma/cm.sup.2.

Detailed Description Text (160):

The cell stack reduced the copper concentration of the solution treated by at least 90 percent. The stack was operated continuously for 600 hours without loss of performance, except for one cell in which a Vexar spacer slipped, apparently during assembly of the cell, which permitted the Tyvek separator 23 (FIG. 1) to contact the platinum screen anode. It was found that the Tyvek separator degraded seriously at those points where it contacted the anode, allowing the cell to short out before the 600 hours operation was achieved.

Detailed Description Text (162):

Another problem to be safeguarded against is that of electrical shorting due to metallic dendrite growth occurring on the cathode and extending toward the anode. Dendrite formation is somewhat unpredictable, depending as it does on the metal ion concentration of the treated solution, the existence of localized high current density paths between the electrodes and, probably, other variables.

Detailed Description Text (163):

In any case, dendrite penetration through the separator elements 23, 23', FIGS. 1 and 3 respectively, must be prevented, as it has been found that electrical short-out dendrites can form in as brief a period as 45 minutes when electrolyzing copper solutions containing from 500 to 1,000 ppm Cu.

Detailed Description Text (164):

As hereinbefore described, Tyvek 1058 has worked well as a separator material, this being a spunbonded sheet formed by the random distribution of very fine translucent continuous fibers (about 0.0002 inch dia.) which are self-bonded by heat and pressure. The composite Tyvek 1058 is about 0.006 inch thick and constitutes approximately 30 fiber layers. Under visual examination, each of these layers presents average triangular openings defined by any three randomly oriented fibers presenting maximum openings measuring 0.0008 to 0.0016 inch. However, since there is a multiplicity of superposed layers, the effective openings for ionic transport are equivalent to devious pores of approximately 0.00016 inch size. Separator structures, as described, have the appearance of translucent sized book paper and are strong enough to prevent dendrite penetration therethrough while still permitting good electrode-to-electrode ionic transport.

Detailed Description Text (165):

In comparison, a Tyvek 1621 sheet, which has been treated to possess openings of about 0.012 inch maximum dimension visible to the unaided eye, i.e., about 100 times larger than the effective size of Tyvek 1058 sheet, did permit dendrite penetration and consequent electrical cell short out.

Detailed Description Paragraph Table (4):

Table 3 _____ Effect of Current on Copper Removal

(starting with an initial Cu conc'n. = 103 ppm.)

		Current	Voltage	Copper Conc'n. (ma)	(volts)
(ppm)		100	2.1	98	200
4.5	67	2,000	6.6	61	

Detailed Description Paragraph Table (7):

		Treatment of Industrial Effluent "B"												
		Pass Current	Voltage	Cu Conc'n.	Efficiency	No.								
(ma)	(volts)	(ppm)	(%)				0	--	--	48.5	--	1	1,000	
3.0	15.5	82	2	500	2.5	5.4	51	3	300	2.3	2.1	55	4	200
											2.2	0.9	30	

Detailed Description Paragraph Table (8):

		Treatment of Industrial Effluent "B"									
		Test No. B-1 B-2									
		Flow Rate, cc/min									
		500 300 Current, ma									
		1500 750									
		Voltage, volts									
		4.0 3.4 Initial iron content, ppm									
		48.5 48.5 Effluent iron content, ppm									
		49.5 49.3 Initial copper content, ppm									
		44.0 44.0 Effluent copper content, ppm									
		18.2 12.5									

Detailed Description Paragraph Table (9):

		Electrolysis of Synthetic Lead Wastes									
		Elapsed Time Lead Concentration (min) (ppm)									
		0	23	4	13.8	8	10.3	12	9.5		

Detailed Description Paragraph Table (10):

		Electrolysis of Synthetic Mercury Waste															
		Elapsed Time Mercury Concentration (min) (ppm)															
		0	30	4	15	8	10	12	5	16	2.5	20	2.0	24	1.5	28	0.7
32	0.5	36	0.4	40	0.3												

Detailed Description Paragraph Table (11):

		Electrolysis of Synthetic Silver Waste														
		Elapsed Time Silver Concentration (min) (ppm)														
		0	20	4	9.8	8	4.2	12	2.6	16	1.7	20	1.0	24	0.8	28
0.7	32	0.6	36	0.5												

Detailed Description Paragraph Table (12):

		Electrolysis of Film Wash Water															
		Elapsed Time Silver Concentration (min) (ppm)															
		0	234	5	95	10	50	15	36	20	30	25	23	30	20	35	13
40	10																

Detailed Description Paragraph Table (13):

		Electrolysis with Foil Electrodes												
		Elapsed Time Copper Concentration Current												
Efficiency (min)	(ppm)	(%)												
			0	105	--	60	67	3.2						
120	42	2.1	180	25	1.4									

Detailed Description Paragraph Table (14):

		MULTI-CELL ELECTROLYSIS										Flow Rate:			
												3.0 gpm			
												Pressure Drop, psi			
												Feed Cell I Cell II Cell III Cell IV Cell V *			
												.DELTA.P.sub.1 **			
												.DELTA.P.sub.5			
												Current Amp. --			
												17 11 8 5 3.5			
												Copper Concentrations			
												Elapsed Time, hrs.			
2	20.9ppm	15.5ppm	11.5ppm	7.7ppm	5.9ppm	4.1ppm	1.25	5.75	4	20.2	16.0	9.1	7.5	4.3	3.0
1.5	6.25	6	19.5	14.2	8.4	6.3	4.0	2.4	2.0	7.0	8	20.6	15.8	11.4	9.3
19.7	14.5	10.3	7.9	5.0	4.0	2.8	8.8	12	20.0	16.5	10.3	8.3	4.1	3.1	3.2
11.5	8.2	4.5	3.1	3.5	10.0										

*Pressure drop across first cell. **Pressure drop across all five cells.

Detailed Description Paragraph Table (15):

Table 14 _____ Decolorization of Industrial Effluent										Elapsed Time from Start Real Dye of Current			
"G" _____										Relative Dye Conc'n. Application Conc'n. of Solution (approximate) (hrs.) in Reservoir			
60' ppm _____										0	1	300	1
0.113 34.2 5 0.051 15.1 6 0.025 7.5 7 0.025 7.5 _____										0.684	205	2	-- -- 3 -- -- 4

Detailed Description Paragraph Table (16):

Table 15 _____ Destruction of Cyanide in Synthetic										Elapsed Time			
Cyanide Waste Experiment No. 1 _____										from CN.sup.- Concentration Start of Current Appl'n. of Solution in mins. Feed Vessel			
60' (ppm) _____										0	145	10	125
116.5 80 115 100 112 _____										20	129	40	117

Detailed Description Paragraph Table (17):

Table 16 _____ Destruction of Cyanide in Synthetic										Elapsed Time			
Cyanide Waste Experiment No. 2 _____										from CN.sup.-Concentration Start of Current Appl'n. of Solution in mins. Feed Vessel			
60' (ppm) _____										--	103	10	101
50 95.7 70 92.5 90 95.6 110 85 130 88.0 150 86.2 _____										20	102	30	103

Detailed Description Paragraph Table (18):

Table 17 _____ Electrolysis of Non-Aqueous										Elapsed Time			
(Dimethylformamide) Solution _____										Copper Concentration mins. ppm			
6 0.6 8 0.3 10 0.2 _____										0	20	2	1.4
										4	0.9		

Detailed Description Paragraph Table (19):

Table 18 _____ (Experiment No. 1)										Elapsed Time, Quinone Conc'n., Hydroquinone			
mins. ppm Conc'n., ppm _____													
40 47.5 56.0 60 34.0 67.7 80 18.5 80.7 100 11.0 108.0 _____										0	97.5	15.0	20
										68.3	36.0		

Detailed Description Paragraph Table (20):

Table 19 _____ (Experiment No. 2)										Elapsed Time, Quinone Conc'n., Hydroquinone			
mins. ppm Conc'n., ppm _____													
40 39.0 80.0 60 19.0 94.0 80 9.8 101.0 100 4.4 106.0 120 2.5 108.0 _____										0	113.0	4.5	20
										68.8	47.0		

CLAIMS:

1. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing provided, at the lower end, with an inlet port for electrolyte introduction, and at the upper end with an outlet port for discharge of said electrolyte, at least one of said electrodes being of uniformly reticulated open construction so as to oppose low resistance to electrolyte flow therethrough and having an extended surface area over which a substantially uniform reaction-producing electrical potential is maintained with respect to surrounding electrolyte throughout the portion of said area in confrontation with the electrolyzing area of the remaining electrode, and said remaining electrode being disposed within an electrically insulator separator envelope closed on all sides except at the edge adjacent to said outlet port fabricated from an electrolyte-inert material which is relatively liquid-tight in construction so as to bar the ready passage of said electrolyte therethrough but which is permeable enough to permit ionic passage between said electrodes.

2. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 1 wherein said separator envelope is fabricated from an electrically insulative web consisting of one of the group made up of ion exchange membranes, textile fabrics, paper and spunbonded products having a pore size barring penetration

of dendrites built up by metal depositing on the cathode electrode of said interfunctioning electrodes.

3. An electrolytic cell according to claim 2 wherein the permeability to said ionic passage is equivalent to a pore size of about 0.2 mil.

4. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 2 wherein said separator envelope is a spunbonded high-density polyethylene having a density of substantially 1.6 oz./yd..² (corresponding to 0.0054 gm/cm.²), and 6 mils (corresponding to 152. μ m) thick, made up of individual fibers in the range of 0.2 mil (corresponding to 5. μ m) diameter.

5. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 1 wherein said uniformly reticulated electrode having an extended surface area over which substantially uniform reaction-producing electrical potential is maintained with respect to said surrounding electrolyte throughout the portion of said area in confrontation with the electrolyzing area of said remaining electrode is a composite made of a plurality of double layers of electrically conductive metal having good corrosion resistance to said electrolyte, each of said double layers being opposite sides of a flattened tubular knit formation fabricated from wire measuring in the range of about 2-4.5 mils (corresponding to a range of about 0.51- 0.115 mm) diameter.

6. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing provided with a separator envelope according to claim 1 wherein said electrodes and said separator envelope are sufficiently flexible to be wound into a layered spiral composite fitting tightly within said leak-tight housing and being closed off centrally and peripherally to bar electrolyte bypass flow around said electrodes and said separator.

7. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 1 wherein there is interposed between said separator envelope and said remaining electrode a relatively thin spacer fabricated from material substantially inert with respect to said electrolyte having a ribbed structure defining full-length open vertical passages confronting said remaining electrode and isolating said separator envelope from contact with said remaining electrode.

8. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 7 wherein said spacer constitutes an extruded netting of high density polyethylene having a thickness of between about 15-30 mils (0.76 mm) made up of about eight strands/inch with said strands intersecting each other in a 90.degree. diamond pattern.

9. An electrolytic cell comprising at least two interfunctioning electrodes disposed at a common transverse level within a vertically oriented leak-tight housing according to claim 1 wherein said electrodes are generally planar in form.